Trends and Health Risks of Dissolved Heavy Metal Pollution in Global River and Lake Water from 1970 to 2017

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Contents

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Abbreviations

CEC	Council of the European Communities
CR	Cancer risk
CSF	Cancer slope factor
EPA	Environmental Protection Agency
EU	European Union
HI	Hazard quotient index
HQ	Hazard quotient
M-K	Mann-Kendall
MLR	Multiple linear regression
PCA	Principal component analysis
RFD	Reference dose
UNECE	United Nations Economic Commission for Europe
US	United States

1 Introduction

In recent decades, heavy metal pollution has become a global environmental issue and the prime focus of environmental security. Such heavy metals derive from both natural sources, such as rock weathering, and anthropogenic sources, such as mining, manufacturing, fertilizer and pesticide use, and waste discharge (Hu et al. 2015; Huang et al. 2015; Ren et al. 2015; Facchinelli et al. 2001; Muhammad et al. 2011). In the last half of the twentieth century, the global amount of heavy metals released to the environment amounted to 22,000 ton of Cd, 939,000 ton of Cu, 783,000 ton of Pb, and 1,350,000 ton of Zn (Singh et al. 2003). Given their solubility, heavy metals can be dispersed by water and, subsequently, contaminate water ecosystems (Nguyen et al. 2005; Jiang et al. 2012; Ağca et al. 2014). In the Buriganga River (Bangladesh), the dissolved metal concentration amounted to 126 μg L^{-1} of Cd, 805 μg L^{-1} of Pb, 5,274 μg L^{-1} of Cr, and 595 μg L^{-1} of As in 2006 (Bhuiyan et al. 2011). Such high levels of heavy metals in surface water pose a direct threat to human health and require urgent attention, as well as further research (Muhammad et al. 2011; Gao et al. 2016).

In an effort to reduce the health risks associated with heavy metal pollution, action has been taken worldwide to control their sources. Since the 1970s, the Congress of the United States (US) has mandated the Federal Environmental Protection Agency (EPA) to regulate the manufacturing, processing, commercial use, labeling, and disposal of such harmful substances (Babich and Stotzky 1985). During the 1980s, the attention of this organization was directed toward regulating the permitted maximum metal concentrations in fertilizers and the maximum metal loading in agricultural land (Mortvedt 1996). In the 1990s, the European Community (now the European Union [EU]) obligated the collection and treatment of municipal

wastewater and prohibited the disposal of wastewater to surface water (CEC 1991). In the 2000s, the Chinese government prohibited the use of leaded gasoline nationwide and issued stricter local emission standards for coal combustion (Duan and Tan 2013).

Measures that successfully control the sources of heavy metal pollution lead to a reduction in the amount of heavy metals released to the environment and, eventually, reduce the metal concentrations in surface water. However, to date, no published research has reported on global levels of heavy metal pollution in water bodies such as rivers and lakes. Therefore, this study collected concentrations of 12 dissolved heavy metals (Cd, Pb, Cr, Zn, Cu, Ni, Mn, Fe, Hg, Al, As, and Co) in global river and lake water bodies from published papers and investigated the trends and health risks from 1970 to 2017 (note, data for the latter four metals were insufficient for analysis). The aim of the study was to explore the sources of heavy metal pollution on decadal and continental scales, to assess the effects of implemented countermeasures for pollution control, and to determine successful measures that could be adopted worldwide.

2 Materials and Methods

2.1 Data Collection

Data on the dissolved concentrations of 12 heavy metal species (Cd, Pb, Cr, Zn, Cu, Ni, Mn, Fe, Hg, Al, As, and Co) in river and lake water worldwide were collected initially from published papers (using a search conducted in Google Scholar and Web of Science). Each sample was assigned a specific year according to the reported sampling date, as follows: for a single sampling year, that year was used; for a sampling range of 1–2 years, the first year was used; and for a sampling range of more than 2 years, the middle year was used. When the sampling date was not provided, the year prior to publication was used. The samples we reviewed had been collected from a total of 120 rivers and 116 lakes in Africa, Asia, Europe, North America, Oceania, and South America and selected from pristine areas, polluted areas, urban area, and estuarine area over the period 1970–2017 (Tables 1, 2, and 3).

2.2 Trend Assessment

In our study, we employed the Mann-Kendall (M-K) test (Mann 1945; Kendall 1975), used extensively to detect change trends in heavy metal pollution over time (Gao et al. 2016; Sharley et al. 2016). As the amount of collected data (number of rivers or lakes) changed by year, the data were classified into five decadal groups (1970–1979, 1980–1989, 1990–1999, 2000–2009, and 2010–2017) to improve the exploration of the trends in dissolved heavy metal pollution in water. The mean

Table 1 Regional distribution of the rivers and lakes considered in this study

Continents	Rivers		Lakes	
	Number	Name of typical rivers	Number	Name of typical lakes
Africa	13[a]	Congo River, Niger River, Nile River, Nyando River, Nzoia River	4[b]	Kainji Lake, Nasser Lake, Victoria Lake
Asia	75[c]	Aras River, Brahmaputra River, Buriganga River, Ganga River, Lean River, Mekong River, Ob River, Pearl River, Pechora River, Tigris River, Yangtze River, Yellow River, Yenisey River	97[d]	Ataturk Dam Lake, Bolgoda Lake, Chaohu Lake, Dongting Lake, Hazar Lake, Hussainsagar Lake, Poyang Lake, Qinghaihu Lake, Taihu Lake, Tasik Chini Lake
Europe	18[e]	Arno River, Danube River, Dordogne River, Elbe River, Mersey River, Rhône River, Stour River, Tiber River	11[f]	Balaton Lake, Hampen Lake, Sortesø Lake, Venice Lagoon
North America	4[g]	Arkansas River, Mississippi River, Tippecanoe River	4[h]	Ivanhoe Lake, Palestine Lake, Thompson Lake
South America	7[i]	Amazon River, Orinoco River, Paraiba do Sul-Guandu River, Pilcomayo River, Sinos River	–	–
Oceania	3[j]	South Alligator River, South Esk River, St. Paul's River	–	–

[a]Dorten et al. (1991), Lalah et al. (2008), Dupré et al. (1996), Banzi et al. (2015), Krika and Krika (2017), and Faton et al. (2015)

[b]Rashed (2001), Muwanga and Barifaijo (2006), and Oyewale and Musa (2006)

[c]Polprasert (1982), Cenci and Martin (2004), Elbaz-Poulichet et al. (1987), Huang et al. (1988), Guay et al. (2010), Martin et al. (1993), Guieu et al. (1996), Dai and Martin (1995), Bradley and Woods (1997), Shiller and Boyle (1987), Cui et al. (2011), Li et al. (2013), Wan et al. (2007), Wang et al. (2018), Varol and Şen (2012), Luo (1984), Sin et al. (1991), Zingde et al. (1988), Shen et al. (1989), Karadede-Akin and Ünlü (2007), Demirak et al. (2006), Karbassi et al. (2008), Kar et al. (2008), Aydinalp et al. (2005), Fan et al. (2008), Turgut (2003), Reza and Singh (2010), Sundaray (2010), Konhauser et al. (1997), Salati and Moore (2010), Varol (2013), Varol et al. (2010), Rahman et al. (2014), Kumar et al. (2013), Li and Zhang (2010), Wu et al. (2002), Zeng et al. (2002), Zhang and Hu (2006), Li and Liu (2009), Cheng and Li (2017), Wang et al. (2015), Li et al. (2008, 2010), Yang et al. (2008), Su et al. (2006), Cheng et al. (2009), Sun et al. (2009), Deng et al. (2016), Qin et al. (2015), Zhang et al. (2016), Gümgüm et al. (1994), Khan et al. (2005), Bhuiyan et al. (2011), Sharma and Vaishnav (2015), Ismail et al. (2013), Rahman et al. (2015), Arefin et al. (2016), Zilkir et al. (2006), Chen and Zhang (1986), Shi (2014), Li (2009), Gong (2011), and Nasehi et al. (2012)

[d]Özmen et al. (2004), Ebrahimpour and Mushrifah (2008), Pathiratne et al. (2009), Barlas et al. (2005), Singare et al. (2010), Jiang et al. (2012), Tao et al. (2012), Rahman et al. (2014), Liu et al. (2010, 2011), Yue et al. (2015), Li et al. (2010, 2013), Wang et al. (2014a, b), Lu et al. (2016), Tian et al. (2011), Yan et al. (2018), Sun and Zang (2012), Mao et al. (2013), Wang et al. (2018), Yang et al. (2008), Sun et al. (2009), Wu et al. (2018), Karadede and Ünlü (2000), Reddy et al. (2012), Farkas et al. (2000), Alhas et al. (2009), Singare et al. (2013), Moore et al. (2009), Zhang (2013), and Meng (2016)

[e]Müller and Förstner (1975), Zwolsman and van Eck (1999), Guieu et al. (1998), Martin et al. (1993, 1994), Dorten et al. (1991), Stoica (1999), Elbaz-Poulichet et al. (1987, 1996), Pettine et al.

(continued)

(1996), Bonanno and Giudice (2010), Bubb and Lester (1994), Say et al. (1981), Adamiec and Helios-Rybicka (2002), and Ramos et al. (1999)
[f]Schierup and Larsen (1981), Martin et al. (1994), Nguyen et al. (2005), and Waara (1992)
[g]Adams et al. (1980), Presley et al. (1980), Martin et al. (1993), Shiller and Boyle (1987), Winner et al. (1980), DeLeon et al. (1986), and Kimball et al. (1995)
[h]Adams et al. (1980), Shephard et al. (1980), Yousef et al. (1984), and McFarlane and Franzin (1978)
[i]Martin et al. (1993), Malm et al. (1988), Shiller and Boyle (1987), Smolders et al. (2003), Hatje et al. (1998), Miller et al. (2004), and Magdaleno et al. (2014)
[j]Thorp and Lake (1973) and Munksgaard and Parry (2001)

dissolved metal concentration in each decadal group was determined as the average of all the collected data in that decadal group. As we inly identified only 92 data points for As, 51 for Co, 38 for Hg, and 7 for Al, these four metals were subsequently removed from the database. M-K tests were conducted on the remaining eight metals (Cd, Pb, Cr, Zn, Cu, Ni, Mn, and Fe) to ensure accuracy of the results (Table 2). We used the M-K calculation methods described by Kisi and Ay (2014) and we used 95% two-tailed confidence levels.

The mean dissolved metal concentrations for each continent were determined as the average of all the collected data for that continent. As the data from Oceania reflected only three rivers (South Alligator River, South Esk River, and St. Paul's River) and there were only three data points for Zn, Cu, and Fe, two data points for Cd and Mn, one data for Pb and Ni, and no data for Cr, this continent was excluded; therefore, only data from Africa, Asia, Europe, North America, and South America were selected to compare the mean dissolved metal concentrations.

2.3 Health Risk Assessment

Humans are exposed to heavy metals via three main pathways: oral ingestion, mouth and nose inhalation, and dermal absorption; ingestion and dermal absorption are the most common pathways for the heavy metal pollution in water (Li and Zhang 2010; Muhammad et al. 2011). The health risk associated with heavy metal toxicity is characterized into non-carcinogenic and carcinogenic. Non-carcinogenic risk, reflected by the hazard quotient index (HI), is defined as the sum of the hazard quotient (HQ) from both exposure routes (oral ingestion and dermal contact). For each exposure route, the HQ is estimated by the average intake of heavy metals from that route divided by the corresponding reference dose (RFD; i.e., the security threshold of a specific metal). When the HI exceeds one, there could be an adverse non-carcinogenic effect on human health. Similarly, carcinogenic risk, reflected by the cancer risk (CR), is the probability of an individual developing any type of cancer over a lifetime and is defined as the sum of CR from both exposure routes. For each exposure route, the CR is assessed as the average intake of heavy metals in that exposure multiplied by the corresponding cancer slope factor (CSF).

Table 2 Dissolved metal concentrations (μg L^{-1}), total sample number (TSN), pristine sample number (PRSN), and polluted sample number (POSN) in global combined river and lake water and Mann-Kendall (MK) test results from the 1970s to 2010s

	1970s				1980s				1990s			
Metals	Mean ± SD	TSN	PRSN	POSN	Mean ± SD	TSN	PRSN	POSN	Mean ± SD	TSN	PRSN	POSN
Cd	9.22 ± 20.61	18	5	15	0.85 ± 2.38	42	14	37	1.05 ± 2.24	29	11	19
Pb	19.13 ± 45.12	7	2	6	11.38 ± 24.48	33	11	28	36.11 ± 84.67	28	11	18
Cr	45.87 ± 71.86	6	0	6	2.28 ± 4.28	11	3	10	9.14 ± 11.51	9	2	7
Zn	233.79 ± 471.83	19	4	17	74.24 ± 230.58	34	11	29	70.81 ± 102.75	23	7	16
Cu	36.51 ± 87.04	17	5	14	4.76 ± 6.11	37	12	32	13.11 ± 32.14	31	11	21
Ni	2.33 ± 3.87	6	2	4	2.76 ± 6.91	17	8	12	45.24 ± 173.48	18	5	14
Mn	165.01 ± 254.78	7	1	7	694.04 ± 607.71	3	0	3	170.89 ± 162.27	9	0	9
Fe	26.97 ± 24.75	8	2	7	222.35 ± 552.27	13	6	10	39.69 ± 81.55	9	2	7
	2000s				2010s				1970–2017			
Metals	Mean ± SD	TSN	PRSN	POSN	Mean ± SD	TSN	PRSN	POSN	MK test	TSN	PRSN	POSN
Cd	10.04 ± 33.77	89	12	88	16.18 ± 52.32	42	11	35	0.24	220	53	194
Pb	26.24 ± 87.07	94	12	92	58.66 ± 113.53	40	12	32	−0.24	202	48	176
Cr	126.14 ± 634.82	85	9	84	130.56 ± 445.50	43	22	25	0.24	154	36	132
Zn	118.42 ± 376.74	111	10	108	482.67 ± 1,384.96	41	12	33	−0.73	228	44	203
Cu	34.14 ± 91.82	114	13	111	62.45 ± 212.25	52	11	43	0.24	251	52	221
Ni	40.32 ± 93.77	74	6	72	201.20 ± 656.54	34	11	24	1.22	149	32	126
Mn	180.17 ± 478.27	70	9	68	137.62 ± 171.88	24	14	12	0.24	113	24	99
Fe	553.70 ± 1,412.86	68	7	66	924.81 ± 1,693.36	28	15	15	0.73	126	32	105

Table 3 Dissolved metal concentrations (μg L^{-1}), total sample number (TSN), pristine sample number (PRSN), and polluted sample number (POSN) in global combined river and lake water of five continents from 1970 to 2017

	Africa				Asia				Europe			
Metals	Mean ± SD	TSN	PRSN	POSN	Mean ± SD	TSN	PRSN	POSN	Mean ± SD	TSN	PRSN	POSN
Cd	3.00 ± 2.93	12	3	11	10.71 ± 38.31	150	43	128	0.62 ± 1.28	34	3	33
Pb	34.05 ± 35.23	16	3	15	36.05 ± 98.45	147	41	123	7.57 ± 15.68	28	3	27
Cr	36.94 ± 68.78	11	4	10	128.04 ± 583.13	125	31	104	7.25 ± 9.63	6	1	6
Zn	59.18 ± 65.21	16	2	16	208.05 ± 784.50	158	35	136	96.07 ± 332.16	28	3	27
Cu	34.17 ± 58.42	15	1	14	37.59 ± 134.33	183	44	157	8.48 ± 15.90	29	3	28
Ni	29.73 ± 39.21	13	1	12	91.62 ± 380.75	111	28	91	3.95 ± 4.34	12	1	12
Mn	517.55 ± 898.01	12	0	12	126.26 ± 269.06	86	22	72	75.82 ± 77.23	5	1	5
Fe	1,240.99 ± 1,897.86	5	1	4	566.34 ± 1,424.80	98	28	79	52.17 ± 110.60	5	0	5
	North America				South America							
Metals	Mean ± SD	TSN	PRSN	POSN	Mean ± SD	TSN	PRSN	POSN				
Cd	3.64 ± 4.92	14	2	13	2.71 ± 5.96	8	2	7				
Pb	24.83 ± 47.07	6	0	6	32.50 ± 48.35	5	1	5				
Cr	26.64 ± 64.29	8	0	8	12.48 ± 15.35	4	1	4				
Zn	196.93 ± 327.95	15	2	14	83.47 ± 130.65	8	2	7				
Cu	38.13 ± 99.97	13	2	12	15.35 ± 31.19	8	2	7				
Ni	5.84 ± 10.37	7	1	6	10.87 ± 23.37	5	1	4				
Mn	377.54 ± 518.82	6	1	6	69.00 ± 76.37	2	0	2				
Fe	31.12 ± 51.32	11	2	10	1,158.21 ± 1,213.62	4	1	4				

The calculation methods for the average intake of metals from oral ingestion or dermal absorption and the relevant parameters (RFD, CSF) used here were described by Li and Zhang (2010) and Gao et al. (2016). The non-carcinogenic risk of eight metals (Cd, Pb, Cr, Zn, Cu, Ni, Mn, and Fe) was estimated relevant to the five decades and five continents by using their corresponding mean concentrations in water (Liu et al. 2015; Gao et al. 2016). Owing to a lack of relevant references for some carcinogenic metals (Cd, Pb, Ni, and Cr), only Pb and Cr were selected to estimate their carcinogenic risk relevant to oral ingestion; their carcinogenic risk relevant to dermal absorption was not assessed (De Miguel et al. 2007; Li and Zhang 2010; Liu et al. 2015).

2.4 Source Apportionment

Principal component analysis (PCA) followed by multiple linear regression (MLR) is a useful method for source apportionment (Yang et al. 2017; Ashayeri et al. 2018; Larsen and Baker 2003). In this study, PCA-MLR was used to determine the contribution percentages of the investigated metal sources to water pollution. First, PCA was employed to represent the total variability of the original metal data in a minimum number of factors; that is, factors with an eigenvalue greater than one were extracted (Loska and Wiechuła 2003). The metal source responsible for each factor could be identified by critically evaluating the factor loadings (Wuana and Okieimen 2011; Järup 2003). Subsequently, MLR was conducted using the standardized PCA scores and the standardized normal deviation of the total dissolved metal concentrations as the independent and dependent variables, respectively. Regression coefficients were applied subsequently to estimate the contribution percentages of the various metal sources.

To compare the changes in the metal pollution sources over time, the potential sources in water were classified into four main types, namely, rock weathering, fertilizer and pesticide use, mining and manufacturing, and waste discharge. Source apportionment of metal pollution in river and lake water was conducted using the SPSS V17.0 software (IBM Corp., Armonk, NY, USA).

3 Results

3.1 Trends of Dissolved Heavy Metal Pollution in Water

The concentrations of the dissolved heavy metals in water differed between the five different time groups over the period 1970–2017 (Table 2 and Fig. 1). Most heavy metal species had the highest dissolved concentrations in the 2010s and the lowest concentrations in the 1970s or 1980s. Collectively, increasing trends were shown in the water for Cd, Cr, Cu, Ni, Mn, and Fe and decreasing trends for Pb and Zn.

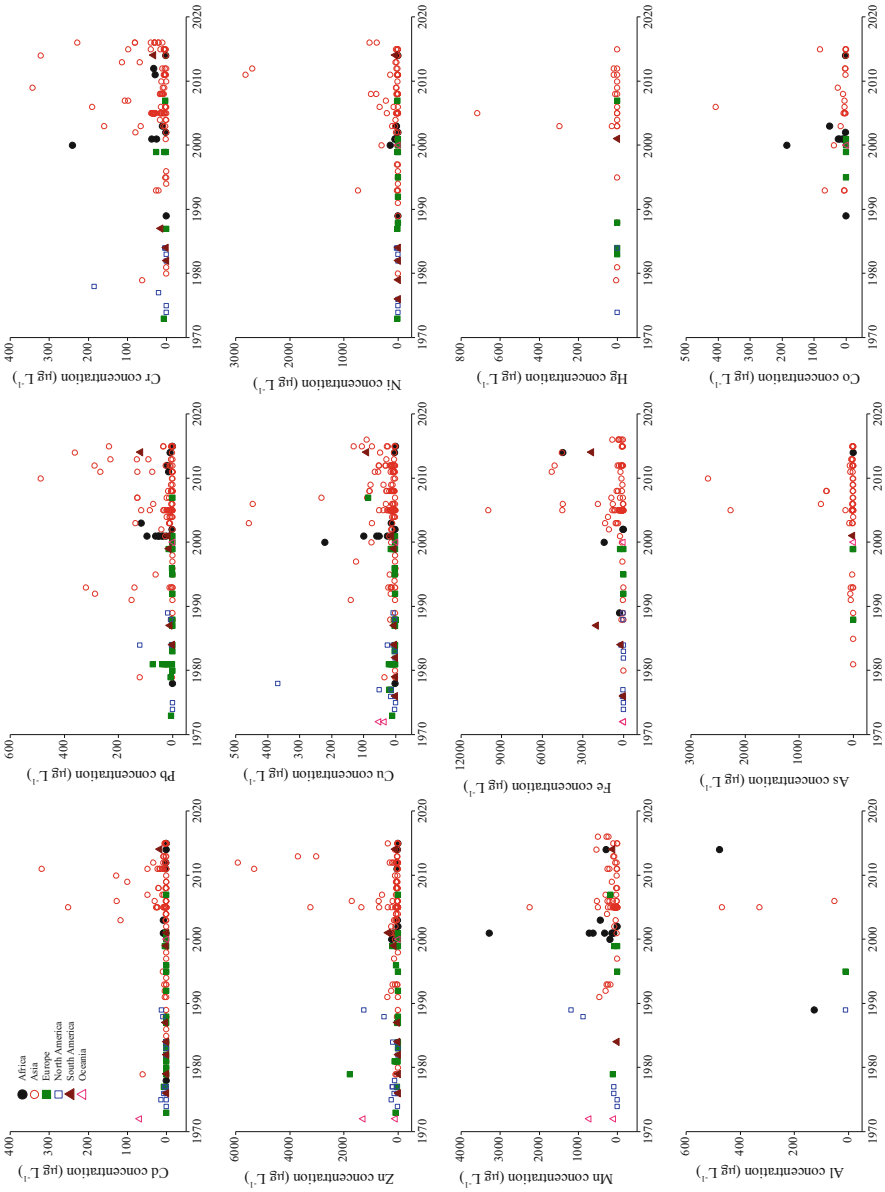

Fig. 1 Dissolved metal concentrations in water by continent from 1970 to 2017 (points represent the mean in one river or lake for 1 year)

The concentrations of dissolved heavy metals in water also differed for the five continents (Table 3 and Fig. 1). Most heavy metal species showed the highest dissolved concentrations in Asia and the lowest in Europe.

3.2 Human Health Risks of Dissolved Heavy Metals in Water

The hazard quotient indices of most heavy metals relevant to non-carcinogenic risks were generally lower than one (Table 4). However, these indices were greater than one for Pb in the 2010s, for Cr in the 2000s and 2010s, and for Zn in the 1970s, 2000s, and 2010s, as well as for Cr in Asia, and for Zn in Asia and North America.

The cancer risks related to Pb contamination were all in the secure range (10^{-6}–10^{-4}) on the five continents over the five decades. However, the cancer risks associated with Cr contamination exceeded the hazardous level in the 1970s, 2000s, and 2010s, as well as in Africa, Asia, and North America for the entire period.

3.3 Sources of Dissolved Heavy Metal Pollution in Water

The main pollution sources in water have changed significantly over time (Table 5). In the 1970s, the main metal sources were fertilizer and pesticide use, along with mining and manufacturing, with a total contribution exceeding 64%. In the 1980s, with the same sources as in the previous decade, the total contribution increased to more than 78%. In the 1990s, the main sources were mining and manufacturing, along with rock weathering, with a total contribution exceeding 58%. In the 2000s, with the main sources remaining the same, the total contribution exceeded 46%. In the 2010s, with the dominant sources the same as those in the 2000s, the combined contribution increased to more than 65%.

Furthermore, the main pollution sources in water differed by continent (Table 6). In Africa, the main metal sources were waste discharge and rock weathering, with a total contribution exceeding 59%. In Asia and South America, the main sources were mining and manufacturing, along with rock weathering, with total contributions exceeding 53% and 60%, respectively. In Europe, mining and manufacturing, waste discharge, and rock weathering were all dominant sources, with a total contribution of 97%. In North America, fertilizer and pesticide use, along with rock weathering, were the main sources, with a total contribution exceeding 86%.

4 Discussion

From 1970 to 2017, river and lake water worldwide showed an increasing trend in the concentrations of dissolved Cd, Cr, Cu, Ni, Mn, and Fe and a decreasing trend for Pb and Zn. This indicated that the heavy metal loadings in water had increased for

Table 4 Non-carcinogenic risk (hazard quotient index) of each metal and carcinogenic risk (cancer risk) of Pb and Cr over five decades and five selected continents

	Decades					Continents				
Metals	1970s	1980s	1990s	2000s	2010s	Africa	Asia	Europe	North America	South America
Cd	5.58E-01	5.14E-02	6.35E-02	6.08E-01	9.79E-01	1.82E-01	6.48E-01	3.75E-02	2.20E-01	1.64E-01
Pb-non cancer	3.75E-01	2.23E-01	7.08E-01	5.14E-01	1.15E+00	6.67E-01	7.07E-01	1.48E-01	4.87E-01	6.37E-01
Cr-non cancer	5.06E-01	2.52E-02	1.01E-01	1.39E+00	1.44E+00	4.08E-01	1.41E+00	8.00E-02	2.94E-01	1.38E-01
Zn	2.14E+00	6.78E-01	6.47E-01	1.08E+00	4.41E+00	5.41E-01	1.90E+00	8.77E-01	1.80E+00	7.62E-01
Cu	2.51E-02	3.27E-03	9.02E-03	2.35E-02	4.30E-02	2.35E-02	2.59E-02	5.83E-03	2.62E-02	1.06E-02
Ni	3.28E-03	3.88E-03	6.36E-02	5.67E-02	2.83E-01	4.18E-02	1.29E-01	5.55E-03	8.21E-03	1.53E-02
Mn	1.91E-01	8.03E-01	1.98E-01	2.08E-01	1.59E-01	5.99E-01	1.46E-01	8.77E-02	4.37E-01	7.98E-02
Fe	1.06E-03	8.73E-03	1.56E-03	2.17E-02	3.63E-02	4.87E-02	2.22E-02	2.05E-03	1.22E-03	4.54E-02
Pb-caner	1.91E-06	1.14E-06	3.60E-06	2.62E-06	5.85E-06	3.40E-06	3.60E-06	7.56E-07	2.48E-06	3.24E-06
Cr-cancer	2.69E-04	1.34E-05	5.37E-05	7.41E-04	7.66E-04	2.17E-04	7.52E-04	4.26E-05	1.56E-04	7.33E-05

Table 5 Rotated component matrix of selected metal concentrations in global combined river and lake water by decade

	1970s			1980s			1990s			2000s			2010s		
Metals	PC1	PC2	PC3	PC1	PC2	PC3	PC1	PC2	PC3	PC1	PC2	PC3	PC1	PC2	PC3
Cd	0.93	−0.03	0.08	0.94	0.15	−0.04	0.82	−0.08	0.02	0.30	0.86	−0.01	0.68	−0.02	0.01
Pb	0.04	−0.24	0.67	0.07	0.89	−0.09	0.78	0.13	0.34	0.88	0.15	0.10	0.21	0.07	0.31
Cr	−0.10	0.89	0.31	−0.13	−0.09	0.71	0.07	0.80	−0.26	0.83	0.13	0.15	0.94	0.05	0.11
Zn	0.56	0.05	−0.04	0.93	0.00	−0.11	−0.01	0.15	0.91	−0.02	0.90	0.18	0.69	−0.03	0.67
Cu	0.10	0.87	−0.08	0.06	0.93	0.05	0.43	−0.11	0.42	0.13	0.12	0.77	−0.14	−0.09	0.96
Ni	0.09	0.28	0.65	0.17	−0.40	0.36	0.83	0.27	−0.32	0.61	0.48	0.10	0.95	0.01	0.09
Mn	0.84	−0.01	0.02	0.92	−0.09	−0.03	0.03	−0.84	−0.10	0.41	−0.01	0.24	−0.02	0.93	−0.08
Fe	−0.08	0.16	0.73	−0.06	0.03	0.86	0.10	0.81	0.26	0.21	0.06	0.81	0.03	0.93	0.08
Eigenvalue	1.98	1.92	1.24	2.73	1.89	1.29	2.52	1.85	1.33	3.10	1.24	1.01	3.03	1.75	1.25
Cumulative percentage	24.72	48.69	64.18	34.18	57.81	73.97	31.50	54.58	71.22	38.80	54.25	66.81	37.84	59.75	75.36
Possible source[a]	FP/ MM	MM/ RW	WD/ RW	FP/ MM	FP/ WD	MM/ RW	WD/ FP	MM/ RW	FP/ RW	MM/ WD	FP	MM/ RW	MM/ WD	MM/ RW	FP/ RW
Contribution percentage	64.17	18.10	17.73	78.14	3.33	18.54	18.97	58.27	22.76	34.60	19.86	45.54	19.44	64.58	15.98

[a]*RW* rock weathering, *FP* fertilizer and pesticide use, *MM* mining and manufacturing, *WD* waste discharge

Table 6 Rotated component matrix of selected metal concentrations in combined river and lake water by continent

	Africa				Asia			Europe			North America			South America	
Metals	PC1	PC2	PC3	PC4	PC1	PC2	PC3	PC1	PC2	PC3	PC1	PC2	PC3	PC1	PC2
Cd	−0.16	0.80	−0.11	0.08	0.70	0.28	−0.02	0.20	0.75	0.22	0.96	0.03	0.11	0.96	−0.02
Pb	−0.04	0.28	0.53	−0.20	0.08	0.89	0.05	0.58	0.33	0.01	0.08	0.13	−0.68	0.89	−0.03
Cr	0.04	0.11	−0.04	0.96	0.16	0.83	0.07	0.66	−0.16	0.42	0.00	0.97	0.09	0.92	0.02
Zn	0.32	0.77	0.10	0.03	0.87	0.12	0.03	0.15	−0.08	−0.64	0.94	−0.01	−0.12	−0.02	0.42
Cu	0.91	0.27	−0.01	−0.18	0.12	0.46	0.03	−0.03	0.82	−0.26	0.00	0.98	0.08	0.94	−0.09
Ni	0.89	−0.18	−0.07	0.26	0.84	0.07	0.06	0.94	0.05	−0.05	0.18	0.10	0.55	0.04	0.92
Mn	−0.05	0.00	0.85	−0.06	−0.02	−0.01	0.93	0.10	−0.11	0.79	0.92	−0.02	0.03	−0.72	0.48
Fe	0.02	−0.29	0.67	0.19	0.08	0.14	0.91	0.76	0.08	−0.17	−0.09	0.26	0.63	0.79	0.41
Eigenvalue	1.90	1.47	1.41	1.07	2.62	1.65	1.24	2.40	1.52	1.13	2.69	2.09	1.10	4.61	1.40
Cumulative percentage	23.77	42.15	59.84	73.18	32.80	53.45	68.98	30.01	49.03	63.13	33.63	59.74	73.55	57.67	75.12
Possible source[a]	MM/RW	FP	WD/RW	MM	FP/WD	MM/WD	MM/RW	MM/WD	FP	MM/RW	FP/RW	MM	WD/RW	FP/WD	MM/RW
Contribution percentage	2.93	21.32	59.15	16.60	22.06	25.18	52.77	48.07	3.13	48.81	85.74	7.40	6.86	40.50	59.50

[a]*RW* rock weathering, *FP* fertilizer and pesticide use, *MM* mining and manufacturing, *WD* waste discharge

the former and decreased for the latter over that period. This finding showed the poor results obtained and illustrated the failure of the measures to control metal pollution. Furthermore, the dissolved heavy metal concentrations differed by continent. As a typical developed region, Europe had the lowest dissolved concentration for most heavy metal species (Cd, Pb, Cr, Cu, Ni, and Mn). In contrast, developing regions such as Asia had the highest dissolved concentration for Cd, Pb, Cr, Zn, and Ni. This indicates not only that the measures to control pollution have been more successful in Europe than in Asia but also that such measures should be extended to other regions.

The health risk assessment showed that the hazard quotient indices of most heavy metal species (Cd, Cu, Ni, Mn, and Fe) were smaller than one, suggesting that these metals posed negligent non-carcinogenic risks to human health. However, the hazard quotient indices of other heavy metal species, such as Pb (in the 2010s), Cr (in the 2000s and 2010s), and Zn (in the 1970s, 2000s, and 2010s), were greater than one, implying that these metals could have caused adverse non-carcinogenic effects to human health in those decades. As regards the two selected carcinogenic heavy metals, the cancer risks associated with Pb from oral ingestion were lower than the hazardous level for the five decades and five continents. However, the cancer risks related to Cr exceeded the hazardous level in the 1970s, 2000s, and 2010s, as well as in Africa, Asia, and North America over the entire period. As exposure to Cr is associated with a high risk of contracting cancer and as there are other pathways (dermal contact and inhalation) to heavy metal exposure in addition to the oral ingestion considered in the present study, preventing such exposure should be a matter of high concern (De Miguel et al. 2007; Li and Zhang 2010; Liu et al. 2015). In addition to water, exposure to heavy metals from the food chain (vegetables, rice, fruits, fish, and other food) could lead to the accumulation of such metals, ultimately leading to chronic toxic effects in humans (Yi et al. 2011; Gao et al. 2016). Therefore, the actual health risks of heavy metal exposure to humans could be more substantial than the results this assessment indicates. Consequently, this should be a matter of global concern.

Source apportionment showed that the main heavy metal sources in river and lake water have changed over time. From the 1970s to the 1980s, these sources were mining and manufacturing, along with fertilizer and pesticide use. A study in 1981–1983 found that mineral refining was the main source for Cd, Hg, Pb, Cu, and Zn pollution in surface water in the National Park of Doñana (Baluja et al. 1985). From the 1990s to the 2010s, the sources were mining and manufacturing, along with rock weathering. A study in 2003 showed that sources of Cd and Pb in Kumho River (Korea) originated from mine discharge (Kim et al. 2010). This finding suggests mining and manufacturing as the critical sources to control global heavy metal pollution in river and lake water. Additionally, the metal sources differed significantly by continent, with waste discharge and rock weathering being dominant in Africa; mining and manufacturing, along with rock weathering being dominant in Asia and South America; fertilizer and pesticide use, along with rock weathering being dominant in North America; and mining and manufacturing, waste discharge, and rock weathering being dominant in Europe. For instance, in the Obuasi mining

area of Ghana, Pb in surface water derives from waste discharge from towns and mine machinery maintenance yards (Armah et al. 2010). In the Yangtze River, heavy metals (Cu, Ni, Fe, Co, and Al) are mainly derived from rock weathering (Wang et al. 2011). Therefore, regional pollution-control measures should concentrate on such region-specific sources.

The decreasing trend in the concentrations of dissolved Pb and Zn in river and lake water indicate that the pollution-control measures for these substances had produced positive effects. For example, from 1994, the US EPA Part 503 rule restricted the ceiling concentration of Pb (300 mg kg^{-1}) and Zn (2,800 mg kg^{-1}) in biosolids applied to the land (Agency 1994). In 1998, the United Nations Economic Commission for Europe (UNECE) agreed the Aarhus Protocol on Heavy Metals to control harmful levels of Pb in Europe (Duan and Tan 2013). In China, after the nationwide prohibition of leaded gasoline in 2000, the Pb pollution level decreased remarkably in cities (Wang et al. 2003). Additionally, having the lowest dissolved metal concentration and the lowest health risk demonstrates the efficiency of the pollution-control measures in Europe; we suggest that these measures should be adopted by the rest of the world. Since the last century, the maximum heavy metal concentration permitted in fertilizers has been regulated in Europe. For instance, in the mid-1990s, the Dutch government proposed regulations limiting the maximum Cd concentration in P fertilizers to 35 mg kg^{-1} (Anon 1989). In addition to the maximum Cd concentration of 100 mg kg^{-1} in P fertilizers, Sweden imposed a tax (30 SEK kg^{-1} Cd) on P fertilizers with Cd concentrations between 5 and 100 mg kg^{-1}. The EU placed limits on the metal concentrations in industrial effluents discharged into the Rhine River in an effort to alleviate the metal pollution in the river (Mortvedt 1996). In 1998, the United Nations Economic Commission for Europe signed the Aarhus Protocol on Heavy Metals to control harmful levels of Cd (Duan and Tan 2013). European legislation prescribes the priority order to be applied to waste treatment, such as prevention, reuse, recycling, other recovery (e.g., energy recovery), and disposal (Kelessidis and Stasinakis 2012). These measures, including implementing rigorous standards on metal emissions, limiting the metals added to products, and pretreating metal-contaminated waste effectively, have controlled heavy metal pollution in rivers and lakes and, we suggest, should be extended worldwide.

5 Conclusions

The present study clearly shows that global river and lake water have increasing trends for Cd, Cr, Cu, Ni, Mn, and Fe and decreasing trends for Pb and Zn over the past period from 1970 to 2017. Most heavy metals had low non-carcinogenic risks over this period. The cancer risks associated with Pb were lower than the hazardous level on the five continents over the five decades, whereas the cancer risks related to Cr exceeded the hazardous level in the 1970s, 2000s, and 2010s, as well as in Africa, Asia, and North America over the entire period. This finding illustrates the failure of

measures to control global metal pollution; the high cancer risks related to Cr on these continents should be a matter of high concern.

Over the past five decades, mining and manufacturing were consistently found to be critical sources of metal pollution in river and lake water. The heavy metal sources differed significantly by continent, with waste discharge and rock weathering dominant in Africa; mining and manufacturing, along with rock weathering dominant in Asia and South America; fertilizer and pesticide use, along with rock weathering dominant in North America; and mining and manufacturing, waste discharge, and rock weathering dominant in Europe. Therefore, regional pollution-control measures should concentrate on such region-specific sources.

The lowest mean dissolved concentrations of most heavy metals in Europe suggest that the countermeasures in the continent have successfully controlled heavy metal pollution. Successful measures include implementing rigorous standards for metal emissions, limiting the metal concentrations in products, and rigorously treating metal-contaminated waste. In addition to such environmental measures, ecological approaches should be considered for heavy metal treatment. For example, after landscape reclamation in catchments, heavy metal inputs to the United Kingdom's Fendrod Lake declined (Blake et al. 2007). Therefore, comprehensive application of environmental and ecological measures should be considered for the remediation of heavy metal-polluted rivers and lakes.

6 Summary

Heavy metal pollution in surface water is a global environmental problem. This study analyzed the trends, health risks, and sources of eight dissolved heavy metal species in river and lake water across five continents (Africa, Asia, Europe, North America, and South America; Oceania was excluded owing to a lack of data) for the period 1970–2017. We wanted to assess the effects of various implemented countermeasures to pollution and to determine those that could be adopted worldwide. Collectively, the water system showed increasing trends for Cd, Cr, Cu, Ni, Mn, and Fe and decreasing trends for Pb and Zn. The mean dissolved concentrations of most heavy metals were highest in Asia and lowest in Europe. Most heavy metals had low non-carcinogenic risks over this period. The cancer risks associated with Pb were lower than the hazardous level on all five continents over the five decades, whereas the cancer risks related to Cr exceeded the hazardous level in the 1970s, 2000s, and 2010s, and in Africa, Asia, and North America over the entire period. Mining and manufacturing were consistently found to be critical sources of metal pollution from 1970 to 2017. However, the heavy metal sources differed significantly by continent, with waste discharge and rock weathering dominant in Africa; mining and manufacturing, along with rock weathering, are dominant in Asia and South America; fertilizer and pesticide use, along with rock weathering, are dominant in North America; and mining and manufacturing, waste discharge, and rock weathering are dominant in Europe. Global trends in the metal loadings in water and in relevant

pollution-control measures suggest that countermeasures in Europe have successfully controlled heavy metal pollution. The successful measures include implementing rigorous standards for metal emissions, limiting the metal concentrations in products, and rigorously treating metal-contaminated waste. Therefore, the measures implemented in Europe should be extended worldwide to treat heavy metal pollution in water.

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